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DETECTION OF CM-244 IN PLUTONIUM-BEARING WASTES AT REPROCESSING FACILITIES

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Detection of Cm-244 in Plutonium-Bearing Wastesat Reprocessing Facilities

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There is concern that small amounts of curium present in reprocessing facility low-activity wastes can interfere with plutonium verification measurement of these wastes. In this paper we will present calculational data showing the effect of curium presence on verification measurements and methods of discriminating between curium and plutonium for certain cases that may be encountered during verification activity.

Introduction

To provide an accurate accounting of plutonium present in the reprocessing facility wastes, we must be concerned with the possible presence of ^{244}Cm in the same wastes. Curium-244 has a very high specific activity, the equivalent ^{240}Pu$_{eff}$ mass of 1 gram of ^{244}Cm is 1.06×10^5 grams. Clearly, a significant bias in the Pu assay of a sample can occur if a measurement does not account for the presence of ^{244}Cm in a sample. In this paper we will present a method for the identification of curium contamination in low-activity wastes and a means of obtaining an independent passive neutron assay of ^{240}Pu and ^{244}Cm for certain cases.

Neutron multiplicity counting provides three distinct observable parameters (singles, doubles and triples) and permits the determination of the three unknowns (α, mass$_{Pu}$, mass$_{Cm}$) in a waste matrix. Other parameters of interest in multiplicity counting can be considered as known values, the leakage multiplication (M_L) of waste drums is unity owing to the small quantity of material present in the matrix, the system efficiency (ε) is treated as a known quantity and confirmed by the add-a-source correction as required.

In the case of wastes free of ^{244}Cm contamination, the analysis allows the operator to insure that there is no Cm present in the wastes and that any observed neutron sources are attributable to Pu. If there is some minor Cm contamination in the wastes the curium and Pu masses can be independently measured and reported. In the case of some first separation cycle wastes, the Cm content may overwhelm the neutron counting to such an extent that the Cm and Pu neutrons cannot be distinguished. If this is the case, historical Cm/Pu ratios may be used to infer the Pu content as is done in high-activity waste streams. In this report, we will examine the level of curium contamination that can be tolerated and produce independent Pu and Cm assy from multiplicity couting.

Simulation

The ability to differentiate Pu and Cm by the multiplicity method has been previously investigated at Los Alamos [1]. These preliminary results indicate that this method may be applicable to low-activity waste matrices. The concept is to exploit the difference between known multiplicity distributions of ^{240}Pu and ^{244}Cm. The method involves the determination of the

doubles-to-triples ratio (D/T) observed from the sample, the D/T for pure ^{240}Pu and ^{244}Cm are known values for a particular counter for a non-multiplying sample. As a small amount of ^{244}Cm is added to the waste the observed D/T ratio is shifted to a lower value due to the higher average number of neutrons emitted per fission for ^{244}Cm. Precise measurement of the D/T ratio of a given sample allows the determination of the presence or absence of curium.

We have used the MCNPX [2] code to examine the multiplicity counting behavior of Pu-Cm mixtures. Figure 1 shows calculated results for trace Cm contamination of a Pu sample in an standard Active-Well Coincidence Counter (AWCC) configured in thermal mode. The conditions used for this analysis were a dilute, non-multiplying homogeneous sample in a AWCC with a 64 μs coincidence gate width and a 4.5 μs predelay setting.
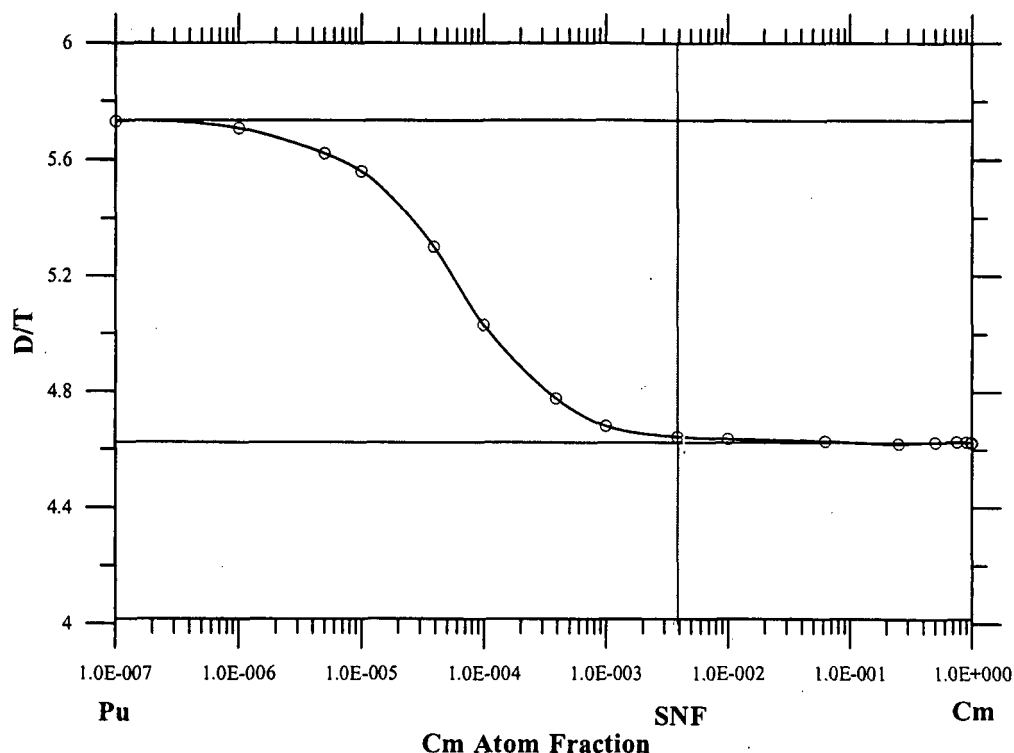

Figure 1. MCNPX simulation of Pu-Cm mixture in an AWCC showing changes in the D/T ratio with Cm concentration.

From Figure 1, it is evident that at ^{244}Cm concentrations above 1 ppm (of spontaneously fissioning species), the D/T ratio is effected and Cm presence can be determined. At ^{244}Cm concentrations above 1000ppm the sample becomes indistinguishable from pure Cm. In the intermediate concentrations it is possible to independently determine the Pu and Cm composition from multiplicity analysis.

Figure 2 shows the result of MCNPX simulation of a high efficiency passive neutron drum counter (A-WDAS). The counter is 37% efficient, has a 64μs coincidence gate and a 1.5μs predelay. In this case Pu and Cm are distributed over the volume of an otherwise empty 200-L waste drum. These data model 2 g ^{240}Pu$_{eff}$ counted for 200 seconds.

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From Figure 2 the curium atomic fraction limits for detection and distinction between species observed for the AWCC simulation are reproduced for the A-WDAS. The values of the D/T ratio are detector-specific and are a function of the system efficiency and coincidence gate fraction for a non-multiplying sample with $\alpha=0$.
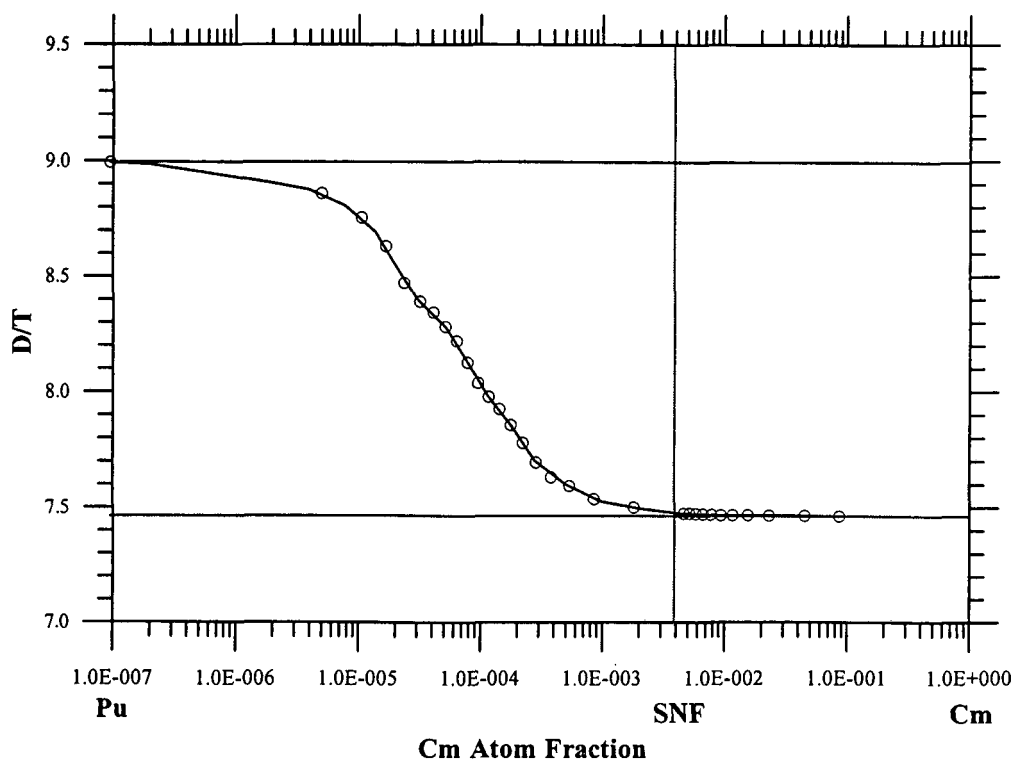

Figure 2. MCNPX simulation of Pu-Cm mixture in an A-WDAS showing changes in the D/T ratio with Cm concentration.

Experimental

We used the five-ring multiplicity counter (5RMC)to try to experimentally observe the effect shown in Figures 1 and 2. The measurements proved rather difficult as the only well-characterized [244]Cm source available for the measurement was a 2mg source. The rather large curium source required the addition of a considerable amount of Pu to obtain the required atom fractions. At the appropriate atomic fractions (5×10^{-5} Cm atoms/atom) the sample multiplication was sufficient to perturb the observed data. The multiplicity distribution for induced fission has a higher average value than for spontaneous fission. We were able to correct for the Pu-only multiplication in the data, but the correction for the fact that the Cm source was driving the Pu samples was not made. No "driver" correction was made because the coupling between the Cm and Pu was not known. That said, the data shown in Figure 3 is partially multiplication corrected multiplicity data from the 53% efficient 5RMC using a 64 µs coincidence gate and a 3 µs predelay setting.
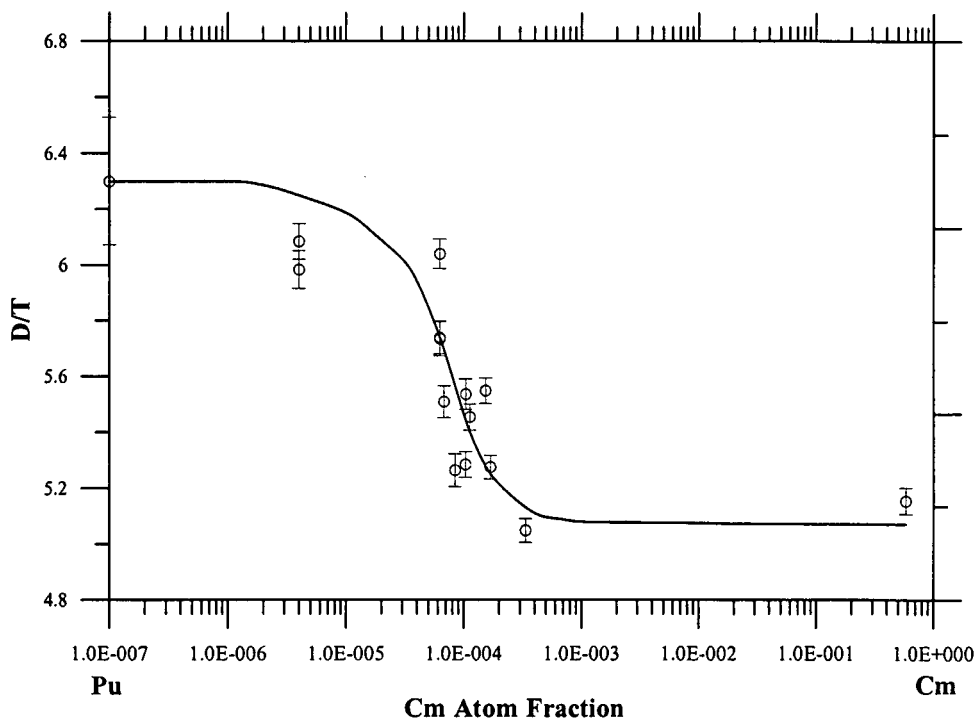
Figure 3. D/T measured in the 5RMC for various Cm dilutions

The data in Figure 3 is overlaid with the trend observed from the simulation data. The statistics in this data set is rather poor, however it bears out as proof-of-principle of the technique. An important observation from this experiment is that this analysis should only be applied to samples with a multiplication of unity.

Conclusions

For curium contamination greater than 1 atom per 10^6 Pu atoms, the presence of Cm in low-activity wastes can be determined by examining the D/T ratio observed from the sample. In the range of 1 to 1000 ppm it is possible to independently quantify both Pu and Cm in sample. Above 1000 ppm Cm the sample is indistinguishable from pure Cm using only this analysis.

The line labeled SNF shown in Figure 1 and 2 is the typical Cm/Pu ratio of spent fuel cooled for 10 years. The ability to quantifiably distinguish between Pu and Cm only exists below the SNF ratio. First separation cycle wastes will typically have Cm concentrations above the SNF ratio and thus will be indistinguishable from pure Cm. And quantification of Pu in these samples is not possible from this technique. However, in the reverse, the analysis method will allow the certification of the absence of Cm in a particular sample originating from the first separation cycle. Wastes generated after the first separation cycle will have Cm contamination below the SNF line and will lend themselves to independent determination of the Pu and Cm mass or the certification of the absence of Pu in the waste.

Despite the limited range of curium contamination where Cm and Pu can be independently determined by this technique, the ability to certify the absence of Cm in a waste drum with only passive neutron measurement is very useful when performing waste measurements from

reprocessing facilities. If the wastes are being assayed for "measured discard" purposes the possibility of a significant bias in the reported result is greatly reduced by the application of this technique.

References

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